OIL PAINTING ON STONE: A CASE STUDY ON ORIGINAL TECHNIQUE AND DETERIORATION OF AN EARLY 20TH CENTURY PAINTING BY GIUSEPPE CALÌ IN MALTA

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Abstract
Maltese wall paintings from the Baroque period onwards were mostly executed in oil or oil-based media applied directly to local limestone (Globigerina Limestone), which is a soft, very porous stone (porosity ~ 40%). These murals are often found in a poor state of conservation. One such painting on a dome of a church located near Malta's main harbour was chosen as a case study. Painted in 1903 and heavily restored in 1963, the painting's advanced state of deterioration makes it an excellent example to review/examine and understand the most common effects and processes of deterioration related to similar wall paintings, and thus to provide essential information for establishing a proper conservation strategy.
Keywords: oil painting on stone, painted limestone, salt weathering, gypsum, nineteenth century painting technique, Globigerina Limestone, Malta

1. Introduction
Malta is the main island composing the Maltese archipelago, located in the Mediterranean roughly 100 km southwest from Sicily. The geology of the islands comprises rocks of sedimentary nature, among which the Globigerina Limestone is a pale yellow biomicritic limestone, fairly homogeneous in texture and colour, with a total porosity around 40% (Cassar 2002). Being soft and easy to quarry has made the Globigerina Limestone the main building material of the Maltese islands since prehistoric times. The ease with which this stone can be carved suited perfectly the richly-decorated Baroque style, so widespread over the Maltese built territory.

Wall painting has played an important role in the interior decoration of Maltese palaces and churches and, at least since the Baroque period, *secco* techniques have been the most diffused mural painting methods. In spite of the general belief that Maltese wall paintings were usually carried out using oil paint, very few scientific studies exist to confirm this statement (Brandi 1951, Liberti and Secchi 1951). This is surprising considering the poor state of conservation in which these paintings are often found.

The present research has focused on identifying materials, techniques and deterioration processes on a wall painting located on the dome of the chapel of the Crucifix in the church of the Immaculate Conception in Cospicua (Malta). This was painted in 1903 by one of the most prolific and popular Maltese artists at the turn of the twentieth century, Giuseppe Calì (1846-1930) (Buhagiar 1987, Fiorentino and Grasso 1991).

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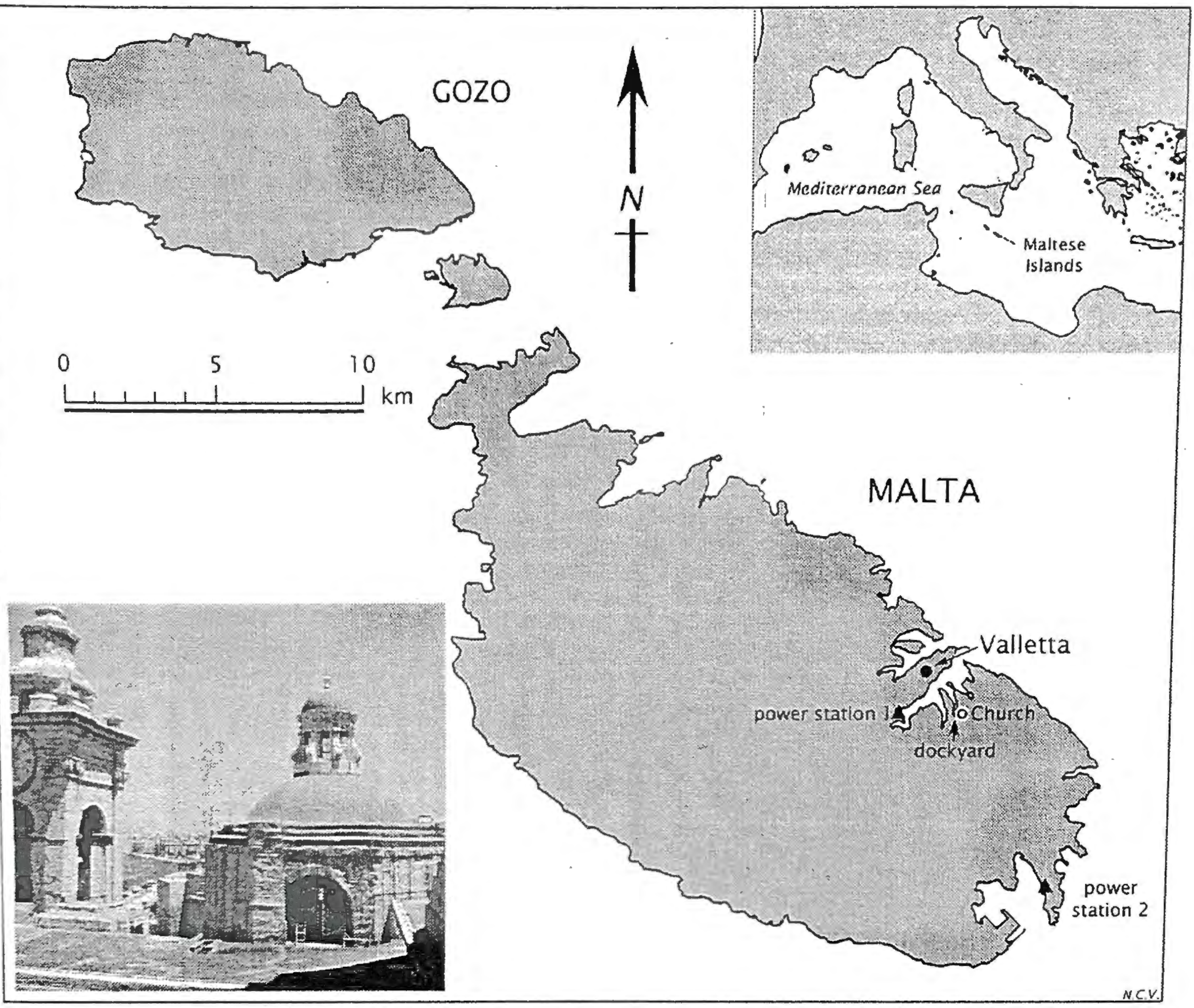

Figure 1: Map of Malta showing the position of the Church, the dockyard and the two power generating stations.

2. The church fabric and the decoration by G. Calì

The church of the Immaculate Conception overlooks a narrow and long bay called Vittoriosa Creek, opposite the capital city of Valletta (fig. 1). The present church fabric is to a large extent the result of substantial modifications dating back to the end of the 17th century, and 1690 is the date by which the chapel of the Crucifix was completed (Galea Scannura 1983, Mahoney 1992). Giuseppe Calì was meant to decorate the entire chapel, however for unknown reasons he eventually painted only the dome and the four pendentives.

After their completion in 1903, the only documented restoration of the dome paintings occurred in 1963. By that time approximately half the painting by G. Calì was already lost and the restorer intervened extensively by infilling, repainting the areas where the painting was lost and overpainting to match his additions. After that date, no other restoration was carried out and the dome underwent a period of neglect. The hydraulic waterproof plaster (*deffun*) covering the external surface of the dome started to deteriorate and water infiltration commenced. The plaster was replaced in 1999 by a waterproofing membrane. The areas where the water infiltration had occurred show severe losses associated with advanced stone powdering and extensive detachment of both stone flakes and paint layers (fig. 2). The portions where the painting by Calì had already been lost and repainted in 1963 show diffuse, thin and well-adhered efflorescences and large areas of paint flaking.

The general condition of the entire cycle looks precarious with the south half being the most affected. A general view is seen in figure 3.

3. Sampling and analysis

Graphic documentation was carried out to record and understand the types and pattern of deterioration, as well as to understand the modifications undergone by the paintings.

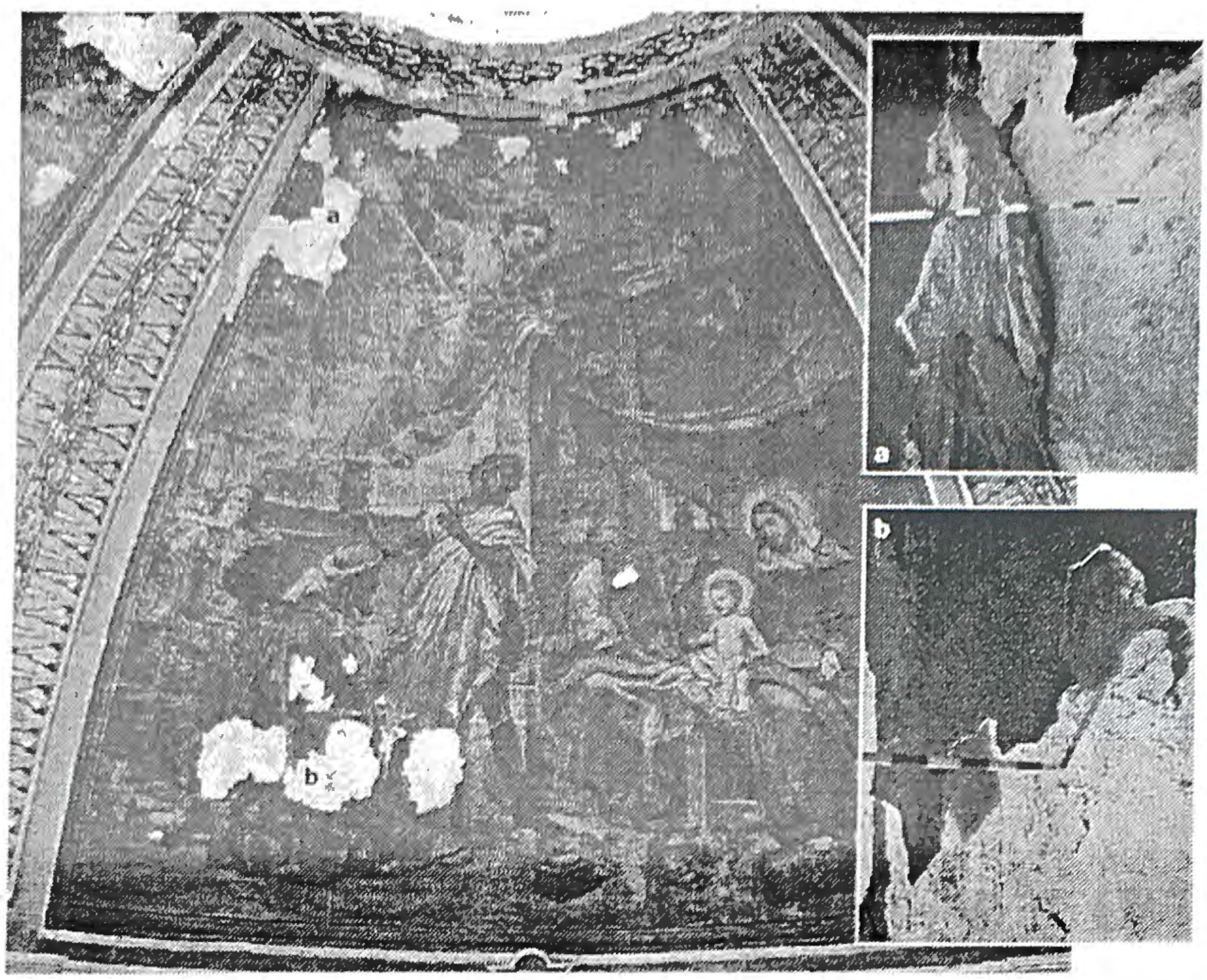

Figure 2: Northwest bay (Adoration) showing two examples of deterioration.

Sampling had two main objectives: to gain an overview of the constituent materials of the paintings, including those applied in 1963 and to identify the agents causing deterioration, namely the soluble salts. The samples taken to identify the constituent materials included stratigraphic painting samples, samples of mortars and plasters, along with two residues of a soft and sticky substance forming drippings on the painted surface. Besides optical microscopy, the analytical techniques used for identifying the inorganic components of the paintings were Scanning Electron Microscopy (SEM) with Energy Dispersive X-ray Spectroscopy (EDS) and X-Ray Diffraction (XRD). The organic components of the painting were identified, after having dissected the samples, using Gas Chromatography (GC) and Pyrolysis-Gas Chromatography (Py-GC), both coupled with Mass Spectrometry (MS). These samples were first prepared according to the combined procedure set up by the Dipartimento di Chimica e Chimica Industriale (DCCI) of the Università di Pisa (Colombini et al. 1999).

To identify type/s and amount of salts causing the decay of the painting, a total of 36 surface samples were taken from the dome both internally and externally. The samples taken inside included stone powder collected from the large stone losses, stone flakes and efflorescences, whilst those taken outside included stone flakes collected from the drum masonry. Powdered samples were also obtained by drilling a hole with a diameter of 5 mm in each of the four bays at approximately 1 m from the drum cornice. The powder produced was collected at three different depths: 0-1 cm, 1-5 cm and 5-10 cm. The profuse dark deposit covering the reliefs on the drum inside the dome was also collected in two areas to identify the airborne particle deposition. All samples were analysed using Ion-Chromatography (IC), whilst the internal surface samples and the dark deposit were also analysed using XRD. SEM-EDS was used for a few samples to observe the morphology of the salt particles and confirm their elemental composition.

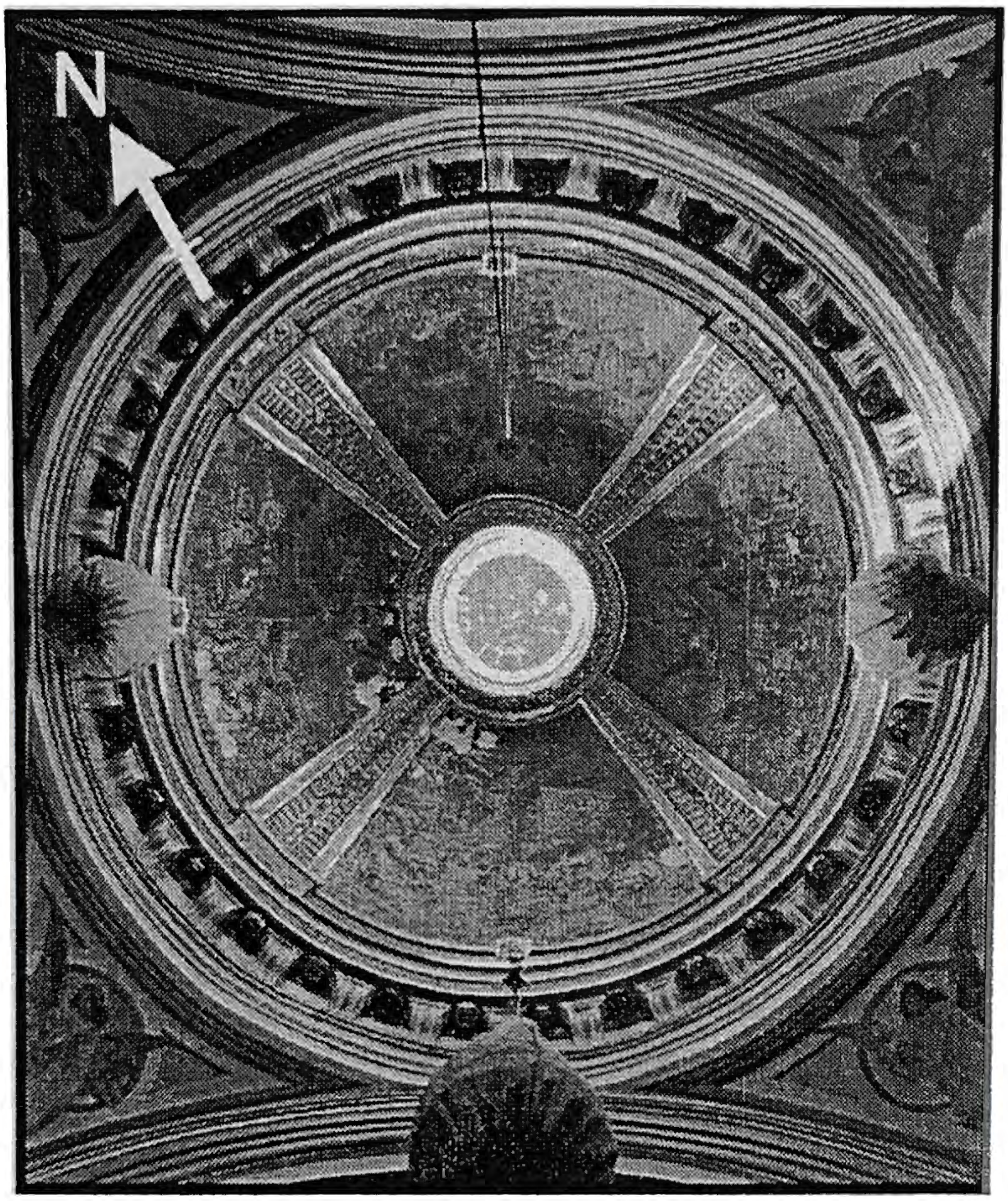

Figure 3: Overall view of the dome.

A 14-month environmental monitoring campaign was also undertaken to understand the role played by the microenvironment in the activation of the salt species present in the painting. Moreover, since the first IC analyses of the stone and salt samples identified high amounts of sulphates, a four-week SO_2 monitoring programme was undertaken in August 2003 using SO_2 diffusion tubes (GRADKO).

4. Results and discussion

4.1 Constituent materials

Cross-sections of the paintings revealed a total of 16 layers. The combination of visual observations, archival research and laboratory analytical techniques demonstrated that the layers roughly belonged to three different periods, including what is being referred to as a pre-Calì period (fig. 4). Some interesting findings, which arose from the stratigraphic and instrumental analyses, included:

- The dome masonry was decorated before 1903 with a painting over the masonry after the interposition of a sealant layer. The binding medium of this paint is egg-based and the pigments analyzed so far were identified as green earth and yellow ochre. The latter consist of coarse and irregular particles, features that might point to a paint produced before the introduction of industrially-manufactured pigments.

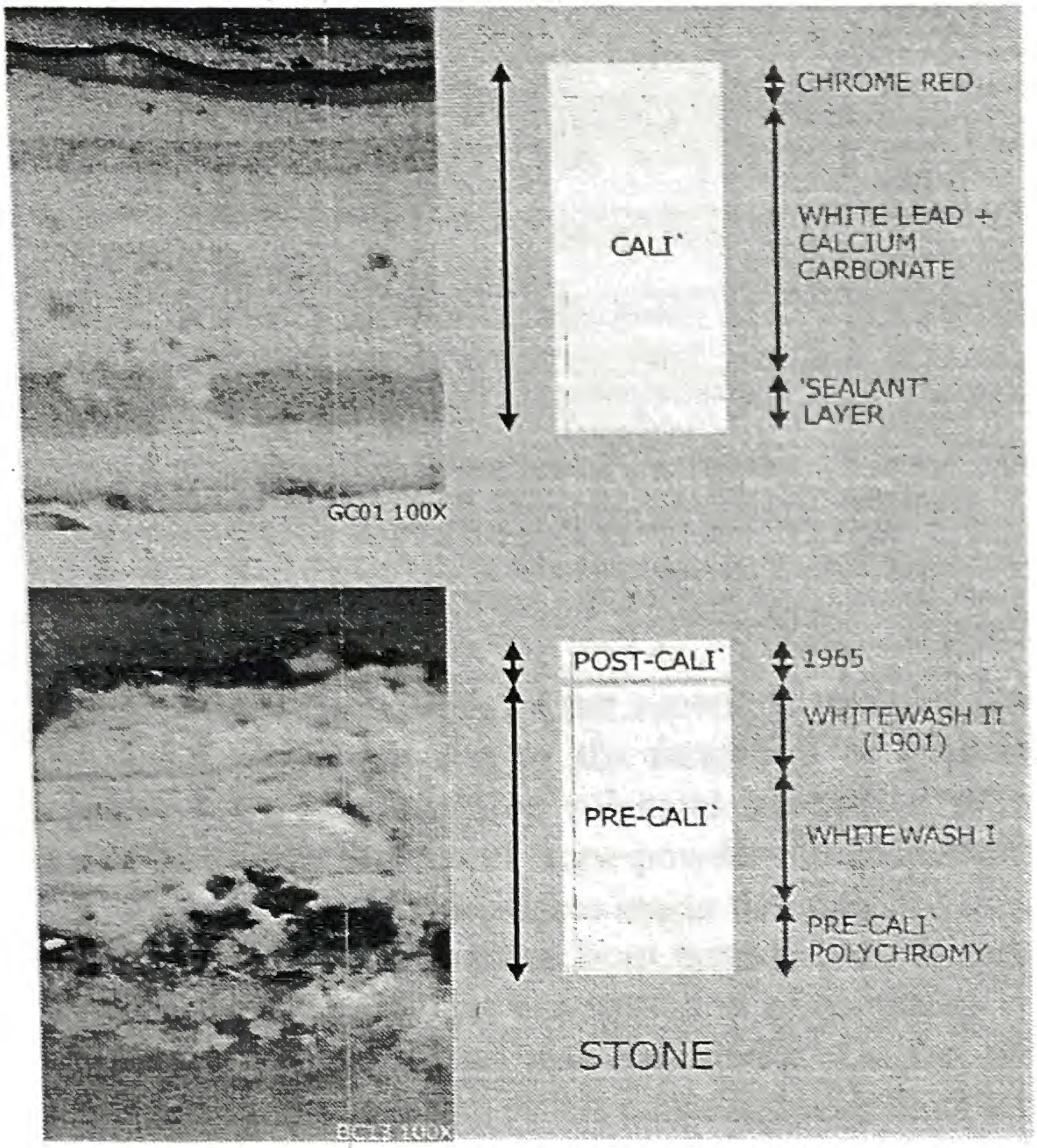

Figure 4: Cross-sections of two samples (GC01 and BC13). These two cross-sections should be seen in sequence, with the exception of the post-Calì layer.

- Over this polychromy, seven or eight layers, mainly composed of white lead bound with linseed oil, were applied during at least two different whitewashing phases (I and II on fig. 4). The last four layers contain an amount of zinc oxide and can be dated back to 1901 thanks to a receipt found in the parish archives listing zinc white as one of the materials purchased then.
- Giuseppe Calì applied about five layers of preparation, characterised by their rough texture, all based on a combination of white lead with smaller proportions of calcium carbonate. GC/MS identified linseed oil with negligible amount of beeswax as binder, along with a quantity of animal glue. The latter may correspond to the sealant used by the artist before laying the white preparation layers.
- The paint layer is well bound to the rest of the preparation. The binder contains linseed oil with negligible amounts of beeswax. Discontinuous traces of varnish are visible on the cross sections, but no resin was detected by GC/MS. The residues found on the painted surface were identified as boiled linseed oil and may be attributed to the 1963 restoration. The range of pigments identified are listed in table 1.
- The 1963 repainting scheme was applied to residual layers of preparation left exposed when the painting by Calì was lost. Pigments were bound with linseed oil, but again very small traces of beeswax were detected by GC/MS along with some animal glue.

- Evidence of pre-Calì repairs was also found and the plaster identified, through both optical and instrumental methods, as a mixture of lime, calcium carbonate with a minor addition of gypsum (or plaster of Paris) (tab. 2, sample GC S 23). This was the only case where gypsum was detected among the constituent materials.

Table 1: Pigments belonging to Calì's palette (1903)

	SAMPLE	PIGMENTS	CHEMICAL FORMULA
RED	GC01	Red ochre; chrome red (lead chromate) and barytes as extender	Fe_2O_3; $PbCrO_4 \cdot Pb(OH)_2$; $BaSO_4$
	GC10	Red lead; vermilion and barytes as base for a red lake	Pb_3O_4; HgS; $BaSO_4$
YELLOW	GC11	Yellow ochre; chrome yellow (lead chromate) and barytes as extender	$Fe_2O_3 \cdot H_2O$; $PbCrO_4$; $BaSO_4$
GREEN	GC12	Chrome-based green; chrome yellow and barytes probably used as extender	Probably CrO_3; $PbCrO_4$; $BaSO_4$
BLUE	GC17	Probably artificial ultramarine and barytes as extender	Probably $Na_{8-10}Al_6Si_6O_{24}S_{2-4}$; $BaSO_4$

4.2 Deterioration products

The data obtained from the analysis of the superficial samples (tab. 2) showed that the most important decay process is due to the formation of gypsum. IC shows that the percentage concentration of extractable sulphates reaches a maximum of 43.5% for efflorescences and 26.2% for samples of stone powder. Drilled samples (figs 5 and 6) show that the concentration of sulphates decreases inside the masonry to a value of around 1%, similar to that found in the samples taken from outside (tab. 2, sample GC S 33). Bay 4 (southeast) is the bay with the highest concentration of sulphates in the range of 0-1 cm, whereas bay 3 (northeast) is the one with the lowest. On the other hand, IC analysis showed that the deposit over the drum reliefs contained high amounts of sulphates (11.5%) and calcium (6.8%), whereas the mineralogical phases found through XRD identified calcite and minor amounts of anglesite (lead sulphate). Gypsum was also identified by SEM in a white deposit found on the back of a detaching stone flake. Surprisingly for a marine environment, chlorides were low with a maximum concentration of 3.3%.

Considering that no gypsum was identified amongst the constituent materials, with the exception of the repair plaster pre-dating 1903, it is plausible to think that an external source of gypsum or sulphur dioxide is responsible for the large amount of gypsum found on the painting. Such salt probably formed as a consequence of the interaction between atmospheric pollutants, introduced by infiltrating water, and limestone. The results of the analysis of SO_2 diffusion tubes positioned inside and outside the dome are still under study, however a preliminary evaluation does not seem to point to high SO_2 levels, in spite of the proximity of the church to the two local power generating stations (fig. 1). The source of SO_2 might have ceased with the closing down of the nearby dockyard, which was located in the creek just below the church. On the other hand, the monitoring of the internal Relative Humidity (RH) and temperature shows a relatively stable and low RH profile.

These paintings are now in a very bad state, one of the main causes being the crystallization of gypsum. The large portions of painting already lost by 1963 and the repair plaster pre-dating 1903, along with the two interventions of whitewashing, seem to emphasize that the fabric has recurrently undergone severe deterioration for a very long time. This fact brings to the fore the intrinsic properties of the Globigerina Limestone and in particular its high porosity (~ 40%) and pore size distribution, which make this stone extremely prone to salt damage (Cassar 2002). Such a tendency is aggravated when the interior masonry decorated with oil painting, or with a low-porosity paint medium, suffers from water infiltration. Once more, the importance of intervening on the fabric to remove any possibility of water infiltration and the need for constant maintenance and monitoring is to be emphasized. Although such needs are important for all materials, this is a must in the case of oil painting on stone, especially when porosity and pore size distribution of the stone support are such that they are prone to salt decay.

Table 2: Results of the salt analysis performed by IC and XRD

IC													XRD			
Sample No.	Bay	Location	Type of sample	extractable amount (%)									C	Gy	H	A
				F^-	Cl^-	NO_3^-	SO_4^{2-}	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}				
GC S 03	1	Int.	E	0.1	1.2	0.6	43.5	1.3	0	0.4	0.1	19.8	++.	++		?
GC S 04	4	Int.	SP	n.d.	0.3	0.3	26.1	0.4	0	0.1	0	13.7	++	++		
GC S 06	4	Int.	SP	0	0.6	0.4	26.2	1.1	0	0.1	0.7	13.8	++	++		
GC S 07	2	Int.	SP	0	0.3	0.3	20	0.5	n.d.	0	0	10.6	++	+		
GC S 11	1	Int.	E	0	1.4	0.4	41.7	1.1	0.1	0.1	0.7	19.6		+++		
GC S 17-I	1	Int.	SP	0	0.3	0.4	22.3	0.4	n.d.	0	0.1	11.3	+++	++		
GC S 23	2	Int.	P	0	0.4	0.4	3.6	0.4	0.1	0	0.1	2.8	++++	+		
GC S 24-I	2	Int.	SP	0	3.3	0.3	0.7	2.9	n.d.	0	0	1.2	++++		+	
GC S 28	2	Int.	E	0	1.2	0.5	16.1	0.9	n.d.	0	0.1	9.3	+++	++		
PW-NE	3-4	Int./Drum	D	0	1.4	1.3	11.5	2.5	0	0.2	0.3	6.8	++			+?
GC S 33		Ext.	F	0	0.3	0.3	0.8	0.4	0.1	0	0	1.2	++++			

Int.=Interior E=Efflorescence P=Plaster F=Flake C=Calcite H=Halite

Ext.=Exterior SP=Stone powder D=Deposit Gy=Gypsum A=Anglesite

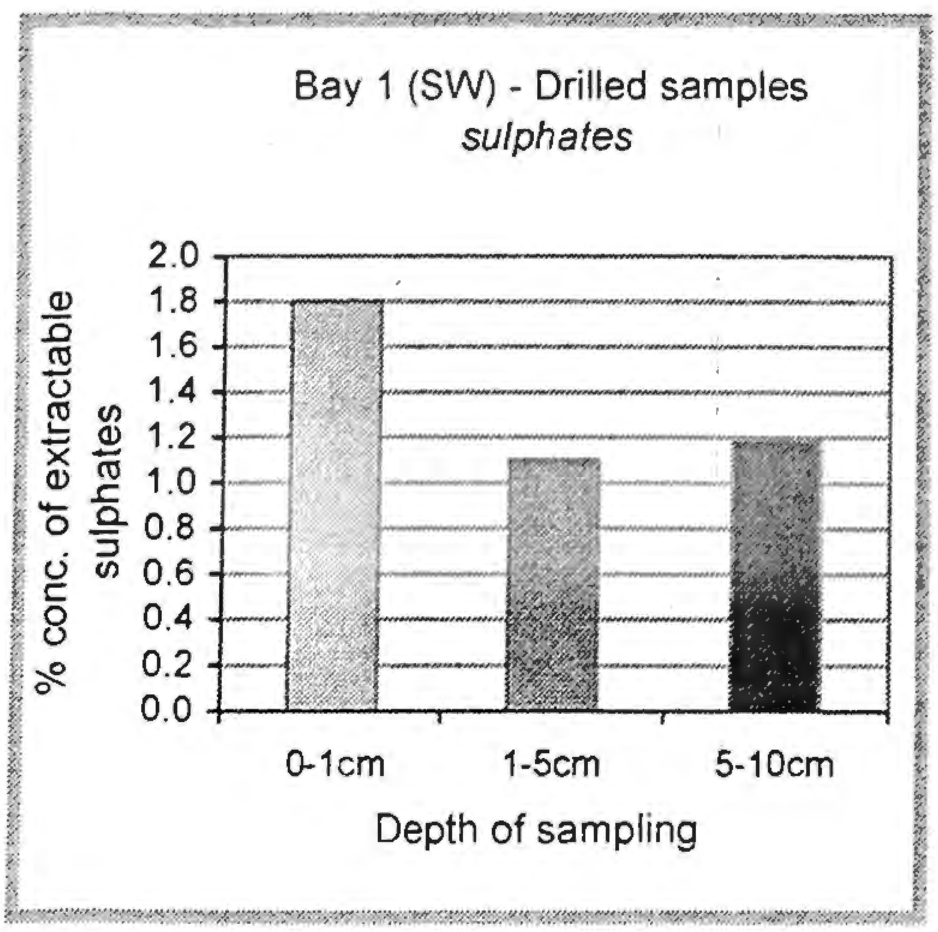

Figure 5: Percentage concentration of extractable sulphates from drilled samples in bay 1 (SW).

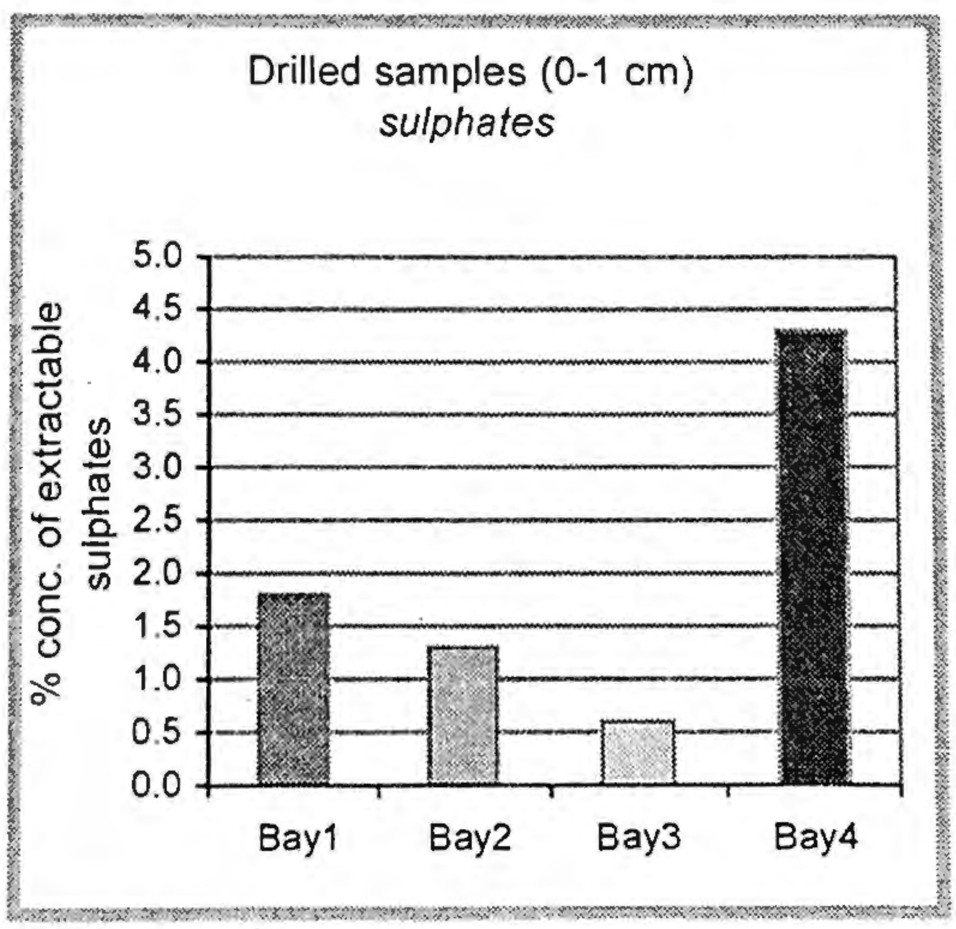

Figure 6: Percentage concentration of extractable sulphates from drilled samples collected at the depth range of 0-1 cm in all four bays.

5. Conclusion

This research programme, which took place over a period of 20 months, has indicated quite clearly that it is the innate composition and properties of both the paintings and the underlying Globigerina Limestone, together with environmental conditions, which are leading to the deterioration of these 20th century paintings. In view of the preservation of the paintings, it is necessary to identify the source of sulphur dioxide, or gypsum, to eliminate it or at least address the problem. On the other hand environmental monitoring seems to indicate that the activation processes leading to decay are over, as long as water infiltration is constantly prevented.

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